Far-Infrared Spectroscopy Group(Annual Report)

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Far-Infrared Spectroscopy Group

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Research Activities

(I) Infrared and Far-Infrared Spectroscopy

a. Design and Construction of a Far-Infrared Spertoscopy System at the UVSOR Facility of Institute for Molecular Science (T. Nanba and M. Ikezawa)

Under the direction of Professor H. Inokuchi of Institute for Molecular Science at Okazaki, we have designed a far-infrared spectroscopy system in collaboration with Prof. M. Watanabe at IMS. Synchrotron radiation from the UVSOR storage ring at the beam line BL6A-1 is reflected by a plane mirror and is collected with a spherical mirror to a spectrometer. The plane mirror is made of copper. The aperture of the collecting system is 80mrad in the horizontal direction and 60mrad in the vertical direction. The collected radiation is collimated and is lead to an interference spectrometer of the Martin-Puplett type, which covers the spectral region from 5mm to 50μm with the resolution of $0.1cm^{-1}$. A higher level of photon fluxes over a conventional source is expected in this spectral region. The transmission and the reflectivity of a small sample is possible because of the high brightness of the light source. The cross-sectional size of the measuring light beam is expected to be as small as $0.5x3mm^2$. The construction of the whole system is now under way.

b. Anisotropic Reflectivity of Ag_2F Crystal (M. Ikezawa, I. Yamamoto and Wang Xiao-lei)

Reflectivity spectra in the near infrared to ultra violet region were observed with polarized light with electric vectors in the directions parallel and perpendicular to the c-axis of the crystal. The spectra are analyzed in an approximate way by the Drude theory. Steep rises of the Drude reflectivity are found at 0.9 and 2.7eV in the respective directions and are interpreted as being due to the anisotropy in the plasma frequency of the free electron. In the spectrum perpendicular to the c-axis, an extremely sharp dip of the reflectivity occurs at 2.9eV. The dip is similar to the "absorption window" of silver metal and is probably due to the cancellation of the dielectric contribution from the free electron with that from the interband transition

of the 4-d electron in the Ag ion.

(II) Alkali Cuprous Halides and Alkali Silver Halides

a. Optical Properties (K. Edamatsu, T. Nanba, T. Awano, I. Yamamoto and M. Ikezawa)

The reflectivity and luminescence excitation spectra of K_2CuCl_3, K_2CuBr_3, Rb_2CuI_3, Rb_2AgBr_3 Rb_2AgI_3, $RbAg_4I_5$ and K Ag_4I_5 crystals were observed at 300, 77 and 15K in the fundamental absorption region from 4 to 30eV at Synchrotron Radiation Laboratory of Institute for Solid State Physics, The University of Tokyo. The analyses of these results are in progress.

b. Band Calculation (K. Edamatsu)

The valence and conduction bands of K_2CuCl_3 and Rb_2AgI_3 crystals were calculated by the self-consistent augmented-plane-wave (APW) method. In both crystals, it has been confirmed that the direct band gap exists at the Γ point in the Brillouin zone. In K_2CuCl_3 the uppermost valence band with about 0.7eV band width has almost d-character of the cupper ions, and the mixing of p-character of the iodine ions is less than 25%. In contrast, the uppermost valence band of Rb_2AgI_3 with about 2.7eV band width has p-character of the iodine ions, and the mixing of d-character of the silver ions is less than 10%. These results are consistent with the UPS and XPS spectra of these crystals. In both crystals, the effective mass of the electron at the bottom of the conduction band shows anisotropy. The effective mass along to the a-, b- and c-axis is 0.41m, 0.41m and 0.36m in K_2CuCl_3, and 0.31m, 0.29m and 0.25m in Rb_2AgI_3. The calculations considering the spin-orbit interaction have been performed by applying the perturbation theory to the valence states at Γ.

c. X-Ray Coloration of Alkali Silver Iodides (T. Awano, K. Edamatsu and M. Ikezawa)

The irradiation of Rb_2AgI_3 and K_2AgI_3 with X-ray at 11K induced anisotropic absorption bands in the visible region from 2.46 to 3.0eV and in the ultra= violet region from 4.10 to 4.37eV. The thermal stability of the absorption bands in the visible region was different from that in the ultra-violet region. When the temperature of the crystal was gradually raised after the irradiation at 11K, the intensities of absorption bands at 2.49 and 2.9eV in Rb_2AgI_3 increased and reached a peak at about 80K and then they decayed above 100K. A similar temperature dependence of the intensity of the 2.52eV band in K_2AgI_3 was observed. On the other hand, the intensities of the absorption bands in the ultraviolet region decreased monotonously above 70K during warming up. This result indicates that the absorption bands in the two regions are ascribed to different defects.

(III) Surface-Enhanced Raman Scattering (SERS) (T. Nanba and I. Yamamoto)

To examine the contribution from the CO molecules in the first monolayer

on the surface and from the overlayered molecules to the SERS process, silver films were prepared by evaporation at 15K in ultra high vacuum and CO molecules were deposit on them. After CO deposition, intense Raman bands occured at 155, 180 and 2140cm^{-1}. The Raman bands were found to disappear above 120K. This result suggests that the chemisorped CO to the silver surface is stable only below 120K.

A third material was put as a spacer between the silver surface and the CO molecules. After covering the first monolayer on the silver surface by Kr atoms as the spacer, about one third of the Raman intensity at 2140cm^{-1} survived compared to that without the spacer.

Overlayered Kr atoms on the chemisorped CO molecules were also found to affect the SERS signal. Further experiments are in progress.

Publication

1) A new radiation sorce for the infrared region, J. Yarwood, T. Shuttleworth, J. B. Hastes and T. Nanba, Nature 312(1984)742.

Master Theses (march 1985)

M1) Radiation Damage Effects in Alkali Silver Halide Crystals, T. Awano.

M2) Raman Scattering from Molecules Adsorbed on Metal Surface, I. Yamamoto.